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LITERATURE SURVEY ON THE EFFECTS OF NEUTRON AND ELECTROMAGNETIC IRRADIATION ON EXPLOSIVES

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U. S. NAVAL ORDNANCE LABORATORY
WHITE OAK, MARYLAND

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LITERATURE SURVEY ON THE EFFECTS OF NEUTRON AND ELECTROMAGNETIC IRRADIATION ON EXPLOSIVES

By

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ABSTRACT: Primary explosives fall in the following order of decreasing stability to gamma radiation: lead styphnate, lead azide, diazodinitrophenol and mercury fulminate. When exposed at the rate of 10^5 roentgen per hour, diazodinitrophenol and mercury fulminate would not perform dependably after doses of 49×10^6 and 32×10^6 roentgen, respectively. No significant change in performance was noted for lead azide and lead styphnate after a total dosage of 52×10^6 roentgen.

High explosives fall in the following order of decreasing stability during gamma irradiation as indicated by gas evolution measurements: TNT, tetryl, RDX, PETN and nitroglycerine. The sensitivity and brisance of TNT and tetryl was not changed significantly by 95×10^6 roentgen irradiation. RDX was shown to be more sensitive after a dosage of 47×10^6 roentgen.

Additional work has been carried out covering the effects of gamma irradiation on some mixed explosive compositions, propellants, primers and detonators.

Most of the work reported to date on neutron irradiation is concerned with primary explosives. Some experiments on lead azide indicate an increase in sensitivity produced by both neutrons and gamma rays and that neutrons are more efficient in causing an increase in sensitivity.

EXPLOSIVES RESEARCH DEPARTMENT
U. S. NAVAL ORDNANCE LABORATORY
White Oak, Silver Spring, Maryland

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At the request of the Bureau of Ordnance the Naval Ordnance Laboratory undertook to determine what had already been done to determine the effects on explosives of exposure to nuclear radiations. This report contains a brief survey and bibliography of all work on this subject so far known to us. The effects of gamma rays on several explosives representing the various levels of thermal stability and sensitivity have been studied and the results are summarized herein. The effects of exposure to slow and fast neutrons have been studied for a few explosives, particulary primary explosives and some work is now in progress at Brookhaven National Laboratory in cooperation with Picatinny Arsenal. This work should be continued and broadened to include more kinds of explosives. The urgency, however, is such that there appears to be little need to enlist the help of other laboratories at this time.

This survey was accomplished under task NO 300-667/76004/01040.

W. W. WILBOURNE
Captain, USN
Commander

J. E. Ablard

J. E. ABLARD
By direction

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I. ELECTROMAGNETIC IRRADIATION

A preliminary investigation of the effects of gamma radiation on explosive materials was carried out jointly between the Los Alamos Scientific Laboratory and the Oak Ridge National Laboratory in 1948 (1). Representative samples of RDX, tetryl, TNT and Comp B were placed in a nest of activated uranium slugs. The radiation received by the samples was of relatively low intensity; during a ten day exposure the total dosage per sample was only 8.6×10^6 roentgen. No visable changes occurred in the samples. Gas produced as a result of this treatment was as follows:

TNT	0.04	cc per gram
Tetryl	0.04	"
RDX	0.14	"
Comp B	0.1	"

The melting points of TNT and tetryl were unchanged while that of Comp B was lowered 0.5°C.

An analogous study was conducted at the Aberdeen Proving Ground (1) where specimens of TNT, tetryl, lead azide, black powder, and propellants M1, M8, and M15 were exposed to 1 Mev X-rays for one hour at a dosage rate of 12 r per sec. The total dose was therefore 4×10^4r. No rise in temperature was observed, and no significant changes in sensitivity were noted.

The most extensive investigation of the effect of gamma radiation on explosives was carried out at the Oak Ridge National Laboratory in conjunction with Picatinny Arsenal (2). Several explosives were subjected to the influence of an intense gamma radiation field at three temperatures: -40°C, ambient, and 71°C. This field was produced by placing the sample in gold-lined magnesium cylinders which were irradiated at one week intervals in a reactor. Reactor irradiation of stable $_{79}Au^{197}$ produces $_{79}Au^{198}$ which decays to $_{80}Hg^{198}$ with a half life of 2.69 days. The decay produces a 0.96 Mev beta and a 0.41 Mev gamma photon.

During irradiation the sample was contained by the tube of a standard vacuum stability apparatus (3) which enabled measurement of the gas evolved. Measurements showed that the glass wall of the sample tube absorbed nearly all of the beta radiation. Weekly readings with a Q842 ionization chamber indicated that the average gamma output from each gold source was 10^5r per hour.

The progress of the decomposition of the explosives was followed by measuring the amount of gas produced. The product gases from some samples then were analysed. Melting points of the samples were measured before and after irradiation.

Impact sensitivities of the samples before and after irradiation were measured on both the Picatinny Arsenal machine and the Bureau of Mines machine. Comparative determinations of the brisance or shattering power of the irradiated and control samples were made with the sand test.

As a result of this work carried out at Oak Ridge it was shown that the primary explosives tested fell in the following order of decreasing stability during gamma irradiation: lead styphnate, lead azide, diazodinitrophenol and mercury fulminate. Diazodinitrophenol and mercury fulminate did not evolve much gas during the first stages of irradiation but the rate of gas evolution increased exponentially. Lead styphnate and lead azide evolved gas at a nearly constant rate. During a total dose of 52 x 10^6r, lead azide evolved 4.9 cc per gram whereas lead styphnate evolved only 0.08 cc per gram. No significant change was found in the impact sensitivity of any of the four compounds after this treatment. No change in brisance was found for lead styphnate or lead azide. However, diazodinitrophenol and mercury fulminate would not perform dependably in the sand test after doses of 49 x 10^6r and 32 x 10^6r, respectively. These doses are therefore sufficient to render diazodinitrophenol and mercury fulminate unreliable as initiators.

The high explosives tested fell in the following order of decreasing stability during gamma irradiation: TNT, tetryl, RDX, PETN, and nitroglycerine. TNT, tetryl and nitroglycerine evolved gas at a nearly constant rate whereas the rate of gas evolution by PETN and RDX increased exponentially. The following table illustrates the wide range of radiation stability observed.

	cc per g after 43 x 10^6r, ambient temperature
TNT	0.08
Tetryl	0.48
RDX	1.49
PETN	2.7
Nitroglycerine	9.0

After exposure to 95 x 10^6r, the melting points of TNT and tetryl were lowered 1.2°C and 1.0°C, respectively. The melting point of PETN was lowered 3.8°C by a dosage of 44 x 10^6r. As indicated by gas evolution and lowering of the melting point tetryl is preferable to PETN as a booster under conditions involving intense gamma fields.

TNT and tetryl became dark colored during irradiation. The color was distributed homogeneously throughout the samples. Nitroglycerine became very viscous and eventually nearly solidified.

The brisance of TNT and tetryl as measured by the sand test was not changed by 95 x 10^6r irradiation. The impact sensitivity height of RDX by the Bureau of Mines machine was lowered from 40 cm to 25 cm after 47 x 10^6r. The minimum drop height required for the Picatinny Arsenal machine was lowered from 9 to 8 inches. No change in the sand test was noted for RDX after this same amount of irradiation.

Tetrytol (tetryl 65%, TNT 35%), standard Baratol ($Ba(NO_3)_2$ 67%, TNT 33%), Composition B (RDX 60%, TNT 40%), Tritonal (TNT 80%, Al 20%), and Pentolite (PETN 50%, TNT 50%) were subjected to gamma radiation at ambient temperatures. The volume of gas formed corresponded closely to the volume formed, under similar conditions, by the major gas-producing component of the mixture except for Pentolite. The rate of gas evolution by Pentolite was irregular and considerably slower than the rate for PETN.

Some of the explosives tested continued to evolve gas after the radiation source was removed. The quantity of gas evolved was sometimes comparable to that evolved during irradiation. In all cases the rate of gas evolution declined after irradiation was terminated. No explanation was offered for this behavior.

All rates of decomposition measured at -40°C were slower than the corresponding rates under ambient conditions, and all rates measured at 71°C were faster. In all cases control runs without irradiation were carried out at 71°C. When samples

were irradiated at 71°C and then allowed to stand at ambient temperature very little post-irradiation gas formation was observed.

Ballistite propellant (69.25% nitrocellulose, 30.30% nitroglycerine, 0.25% diphenylamine) evolved 2.80 cc of gas per gram of sample after irradiation for 38 days at ambient temperature. M15 propellant (54.7% nitroguanidine, 20.0% nitrocellulose, 19.0% nitroglycerine, 6.0% ethyl centralite, 0.3% cryolite) evolved 3.3 cc of gas after 60 days irradiation at ambient temperature. The rate of gas evolution by the M15 propellant did not increase as rapidly as that of ballistite. M15 is much more stable to gamma radiation than ballistite.

M26 primers in groups of one hundred were exposed to gamma radiation for 15, 30, 60, and 90 days at ambient temperature. The M26 primer contains 100 mg of PA 100 primer composition consisting of 53% potassium chlorate, 17% antimony sulfide, 25% lead thiocyanate, and 5% lead azide. The three groups of primers which had been exposed for 15, 30, and 60 days respectively functioned normally in a standard steel ball impact test. Of the fourth group of 100 primers irradiated for 90 days, 98 functioned normally and two burned but failed to explode. Run down tests on sensitivity to stab action in which the steel ball was dropped from lower heights were performed on unirradiated primers and primers exposed for 30 days. The sensitivity to stab action was increased by irradiation.

M19A2 detonators were irradiated and tested in a manner similar to the primers. This detonator contains 105 mg of PA primer composition followed by 250 mg of lead azide and 142 mg of tetryl. Those irradiated for 15 days functioned normally but irradiation for 30, 60, and 90 days resulted in malfunction of 12, 60, and 92%, respectively. The observed malfunctioning consisted of incomplete detonation in which the priming mixture exploded but failed to initiate the adjacent tetryl charge. This malfunction is attributable to failure of the deteriorated lead azide to initiate the tetryl.

The effects of cobalt 60 gamma radiation on a ballistite, JPN type N-2, have been studied by the Naval Powder Factory using the sources at the Brookhaven National Laboratory (4). Strips of the propellant were subjected to total doses of 1, 2, 3, 4, and 5 x 10^6r at an intensity of 1.93 x 10^5r per hour after which they were chemically analyzed and physically tested. Significant changes were observed in the viscosity of the nitrocellulose, the amount of available centralite stabilizer, and the stability of the propellant. Results from an oxygen Taliani test indicate that an exposure of 5 x 10^6r produces an effect similar to an accelerated aging treatment of 20 days at 80°C.

Irradiation produced no significant change in the closed-bomb or strand-burning properties of JPN propellant.

The effect of high energy X-rays on lead azide has been studied at the Armament Research and Development Establishment, Fort Halstead, England (5). Samples of Service lead azide were exposed to the radiation from a million volt generator. The times of heating at different temperatures required to produce detonation of these samples were compared with those of unirradiated samples. It was found that the irradiation produced no effect below dosages of 10^6r, but marked sensitization was produced by a dosage of 7×10^6r.

A reduction of time to detonation of lead azide at elevated temperatures similar to that described above has been observed after irradiation with sunlight and with ultraviolet light (6)(7). A similar sensitization also has been produced by a period of pre-heating insufficient to give detonation (8).

II. NEUTRON IRRADIATION

Studies of the effect of neutron irradiation on explosives have not been as extensive as those concerning gamma irradiation.

The effect of slow neutrons on lead azide has been studied by Bowden and Singh at the University of Cambridge (9). High energy neutrons were produced by deuteron bombardment of a beryllium target in a cyclotron. The neutrons were then slowed down by 20 cm of paraffin wax before striking the sample. During irradiation, the sample was contained by a silica tube in an atmosphere of dry nitrogen at a pressure of 10 cm of mercury. The neutron flux was measured by determining the amount of radioactivity produced in a silver azide sample placed in an adjacent tube. After irradiation for one hour at a flux of 2×10^7 neutrons per cm^2 per sec, the sample did not explode. Its thermal stability was, however, somewhat lower than the unirradiated material as shown by its rate of gas evolution at 315°C.

Lead azide was also bombarded with fission fragments by coating the crystals with a thin film of uranium oxide and then irradiating with slow neutrons (9). Even when bombardment was carried out at 290°C, no explosion took place.

Muraour and Ertaud (10) exposed several primary explosives to neutrons in the pile of Chatillon (Zoé). Lead azide did not explode although exposed to a total neutron dose of 3×10^{14} neutrons per cm^2. Mercury fulminate, lead styphnate, tetracene, and metanitrophenyldiazonium perchlorate were exposed to similar doses. Tetracene turned brown, but no explosions occurred.

Groups of detonators containing PETN were exposed to nuclear radiation in two ports of the "water boiler" reactor at Los Alamos (11). One group was exposed for 3 hours to the following flux:

	Neutrons per cm^2 per sec.
1.4 Mev Neutrons	3×10^9
dE/E Neutrons	9×10^9
Thermal Neutrons	shielded out
Gamma Rays	1×10^{11} (40 r/sec).

Another group was exposed for 10 hours to the following flux:

	Neutrons per cm^2 per sec.
1.4 Mev Neutrons	8.7×10^{10}
dE/E Neutrons	5.5×10^{10}
Thermal Neutrons	2×10^{11}
Gamma Rays	1×10^{12} (500 r/sec).

In both cases the threshold firing voltages of the detonators were unaffected by irradiation.

A study of the effects of reactor radiation on explosives is being carried out by Picatinny Arsenal and the Brookhaven National Laboratory (12). A method has been devised whereby 6 grams of explosive can be irradiated in the Brookhaven reactor without endangering its function should an explosion take place. An increase in impact sensitivity of lead azide was observed after exposure to reactor radiation for 5 days. This effect was more pronounced after exposure for 20 days. The impact sensitivity was increased more by 5 days of reactor irradiation than by exposure to 2.5×10^6r of gamma radiation from a cobalt 60 source. Neutrons are, therefore, more efficient in causing this increase in sensitivity than are gamma photons. These conclusions are based upon results obtained with the Picatinny Arsenal impact machine.

It was also found that reactor irradiation lowered the explosion temperature of lead azide. A five hour exposure lowered the induction time temperature curve about 20-30°C.

It has been found that the effect of radiation can be followed spectrophotometrically by an ultraviolet reflectance method. Infrared is expected to give quantitative data on absorption properties. It is hoped that these spectrophotometric properties can be correlated with explosive characteristics such as sensitivity build-up time and detonation velocity. For this purpose bombproofs have been built with provision for handling radioactive explosives. The studies are being extended to other primary explosives and to standard high explosives.

III. DISCUSSION

Experimental work to date shows that the military high explosives are quite resistant to gamma radiation. The effects on TNT, tetryl and RDX are relatively minor considering the large dosages to which they have been exposed. These three explosives have been subjected to radiation at least 2×10^4 times as great as the maximum permissible for exposed personnel. In addition, the explosives have been exposed continuously for periods up to 90 days in the case of TNT and tetryl while the maximum permissible for personnel is based on an 8-hour day of a 40-hour working week. Of the primary explosives, lead styphnate shows about the same degree of resistance to gamma radiation as the high explosives mentioned above.

It might be of interest to speculate whether or not there would be any appreciable interaction between a typical high explosive placed in the proximity of a reactor such as might occur in the case of a nuclear powered aircraft carrying explosive weapons or devices. Let us use a 10^4 horsepower reactor and a distance of 5 meters from the source of radiation as an example. Under such conditions the gamma radiation would be about 2×10^3 times greater than that to which the high explosives have been exposed and about 4×10^7 times greater than the maximum permissible for personnel. The estimate of gamma radiation in the above example does not take into account absorption of radiation either by materials of construction or shields designed for protection. It appears that a 10^4 horsepower reactor would have to be shielded to protect explosives from damage by radiation. Protecting personnel is by far the greatest problem.

Very little work has been carried out on the effect of neutron irradiation on high explosives. In one experiment at Los Alamos PETN was exposed for 10 hours to a neutron flux nearly equivalent to the estimated neutron flux 5 meters from a 10^4

horsepower reactor. The PETN was not adversely affected in this experiment in which 1.4 Mev neutrons were used. It is estimated that the neutron flux is about 10^9 times greater than the maximum permissible for exposed personnel. If the neutron energy used in the above experiment were converted to heat, not over 2 gram-calories would be formed per hour per cc of explosive. It would appear that exposure of explosives to fast neutrons would require shielding mainly for personnel protection.

Additional work is needed to investigate possible interaction of fast neutrons with high explosives.

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